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FLUID MIXING APPARATUS

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Inventor(s): JUHAS DAVID G; TINNEY JOHN R

Applicant(s): EASTMAN KODAK CO (US)

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Abstract

A fluid mixing apparatus and method are described. Mixing is effected by means of Taylor vortices induced in component liquids while they are in the apparatus mixing zone. Bubble flashing in the mixing region is prevented by use of a smooth-surfaced rotor and by maintaining the Taylor number in the region at or below 340. Adequate and efficient mixing is achieved by maintaining the Taylor number at or above 125. Material waste during cleaning is minimized and a uniform fluid residence time is achieved by maintaining a uniform cross-sectional area throughout the mixing region.

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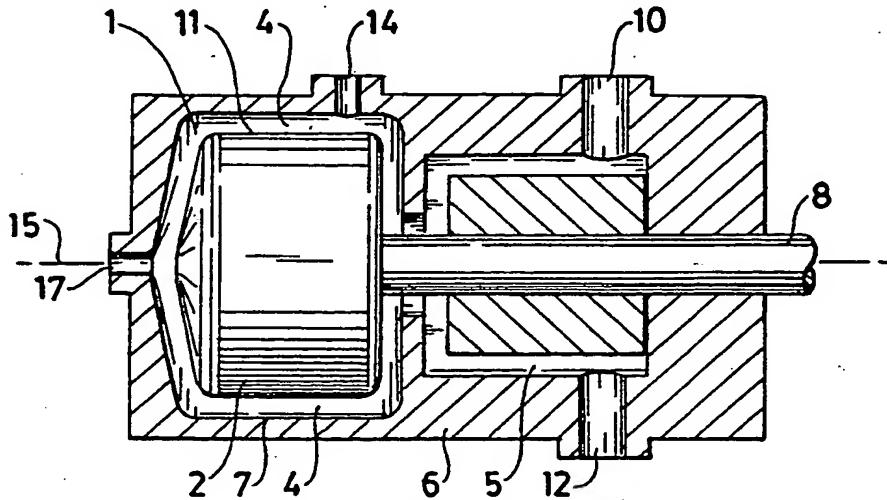
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(54) Title: FLUID MIXING APPARATUS



(57) Abstract

A fluid mixing apparatus and method are described. Mixing is effected by means of Taylor vortices induced in component liquids while they are in the apparatus mixing zone. Bubble flashing in the mixing region is prevented by use of a smooth-surfaced rotor and by maintaining the Taylor number in the region at or below 340. Adequate and efficient mixing is achieved by maintaining the Taylor number at or above 125. Material waste during cleaning is minimized and a uniform fluid residence time is achieved by maintaining a uniform cross-sectional area throughout the mixing region.

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FLUID MIXING APPARATUS

BACKGROUND OF THE INVENTION

5 Field of the Invention

This invention relates generally to fluid mixing, and more particularly, to a method and apparatus for mixing fluids in line by use of Taylor vortices.

10 Background

Conventional mixing apparatus for the continuous mixing of two or more fluids in in-line systems usually make use of rotary blades, spiral ribbons or paddles to mix input liquids in bowl-like or tubular containers.

15 Such apparatus, however, often provide relatively low quality product mixes at low production rates. Another problem with these mixers is that the geometric configuration of the impeller and mixing region sometimes results in cavitation and flashing of gasses from the 20 mixed solution. Although not a problem for many mixing operations, the presence of gas bubbles is undesirable for coating photographic emulsions. Further, the blade/paddle design of prior mixers results in back-mixing, which is undesirable if uniform plug flow mixing is desired, as it 25 is in the preparation of photographic emulsions. Also, adequate cleaning of mixing apparatus with such configurations is both expensive and time consuming.

Another problem with conventional mixers is that they tend to have large hold-up volumes and a number of 30 "dead zones," i.e., non-flow regions, where fluids tend to stagnate. Both of these characteristics make conventional mixers unsuitable for use in mixing photographic emulsions. In the manufacture of photographic film, the same mixing apparatus often are used to process different

chemicals at different times. Before the mixer can begin mixing different chemicals, however, it must be cleaned thoroughly. To avoid inefficiencies resulting from mixer down-time during cleaning, the mixer must be capable of quick and easy cleaning. Also, to avoid wasting chemicals, the mixer should have a small hold-up volume so that the amount of chemicals remaining in the mixer after it completes a mixing operation is minimal. The high cost of such materials makes a large hold-up volume disadvantageous because of the resultant waste of materials when the mixer is emptied for cleaning.

A further problem associated with dead zones is that they result in varying fluid residence times. Because of the importance of a uniform mixer residence time for each discrete unit of fluid, conventional mixers, with their numerous dead zones, are unsuitable for use in mixing photographic emulsions.

A further problem with conventional mixers is the difficulty with purging air upon start up. In the preparation of photographic emulsions, the same mixing apparatus may be used for the production of several different types of emulsion in a single day. Therefore, to reduce inefficiencies resulting from equipment downtime, a mixer must be capable of being changed over for operation on a new set fluids as quickly as possible. One problem with prior mixers is that their geometry causes delay because of the difficulty of purging air from the mixer when it is initially filled. Therefore, a need has arisen for a mixer which allows quick purging of air during start up.

Furthermore, conventional mixers, under certain conditions are unacceptable for mixing photographic materials because they result in an excessive back mixing ratio. Back mixing ratio is defined as the ratio of

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actual fluid residence time in the gap to theoretical residence time. A suitable back mixing ratio in the mixing of catalyzing agents with photographic materials depends on the reaction rate, but generally a back mixing ratio of no greater than 1.2 will be acceptable.

Also, the non-uniformity of gap width in conventional mixers makes plug flow impossible. Plug flow occurs when each unit of fluid entering the mixing zone has the same residence time within the zone. In in-line photographic emulsion delivery systems, it is imperative that plug flow be achievable so that each unit of fluid is mixed uniformly. Plug flow also results in the efficient purging of air and other fluids from the mixer on start-up. Conventional mixers, such as the one described by Suh, et al., are unable to accomplish this because of non-uniform gap width in the mixing region and the presence of dead zones, where fluids tend to stagnate.

Also, conventional mixers can be relatively expensive to manufacture because of the requirements that the rotor operate at speeds of up to 15,000 revolutions per minute (RPM). To achieve the necessary rotational speeds required with the mixer special, relatively expensive, high-speed drive motors must be used. Each of these requirements results in added expense of the mixer. Additionally, the mixer described in Suh further requires the additional expense associated with the use of a destabilizing electrical force to effect suitable mixing.

The structures of many conventional mixers include conical or cylindrical chambers which use correspondingly shaped rotating inner elements having various forms of projections on the surfaces thereof, the components to be mixed being introduced in the gap between the outer surface of the inner member and the inner surface of the outer chamber. The gap is relatively small

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in comparison to the radial dimensions of such members, but the projections result in the undesirable generation of bubbles due to turbulent flow in the mixing chamber. Moreover, the conically shaped chambers and inner members thereof are particularly difficult to construct and to clean so that the cost of manufacture and of operation are relatively high for some applications.

It is desirable to accomplish mixing of photographic emulsions by laminar mixing techniques. Laminar mixing between one outer stationary cylinder and one inner rotating cylinder occurs when the Taylor number characterizing the flow of the fluid in the mixing region is low. Taylor number, T_a , is defined as:

$$15 \quad T_a = (2\pi R_i N d / 60 v) (d/R_i)^{1/2}$$

where N is the inner cylinder (rotor) speed in RPM, R_i is the radius of the inner cylinder radius, d is the mixing zone gap width and v is the kinematic viscosity of the resultant mixture. A Taylor number from about 41.3 to 400 insures laminar flow with Taylor vortices in the mixing region. Laminar mixing is desirable since no bubble flashing occurs and because it is a more energy efficient method of mixing.

Effective laminar mixing is promoted by creating vortex-like motions of the fluids in the mixing region. These laminar vortices, known as Taylor vortices, increase the surface area of the interface between the liquids in the mixing region. Because mixing efficiency increases proportionally with an increased interface surface area, creating Taylor vortices results in more effective mixing per unit of power consumed. Although the use of mixing apparatus employing Taylor vortex mixing techniques is known, no apparatus or method is presently known which can

mix liquids of moderate viscosity by use of Taylor vortices, can prevent the formation of bubbles in the mixed solution and which has a small hold-up volume.

5 BRIEF SUMMARY OF THE INVENTION

In accordance with the invention, an effective high quality mixing of multi-component fluids can be provided at relatively low cost, which mixing can be accomplished with a residence mixing time which is shorter and more uniform than that provided by any presently known commercial apparatus or method. The apparatus of the present invention is also relatively simple and inexpensive to manufacture and is capable of providing non-turbulent mixing of fluids having a wide range of viscosities, including low and moderate viscosity, without the use of a destabilizing force, while also maintaining plug flow.

One embodiment of the present invention provides a Taylor vortex mixer for mixing one or more liquids. The apparatus includes a stationary chamber. The chamber has a substantially cylindrical central wall and a substantially conical end wall. A rotor is positioned within the chamber and it includes a substantially cylindrical central portion, a substantially conical end portion, a central radius, and a longitudinal axis. The rotor also has an outer surface spaced from the walls of the chamber and defining a mixing region of uniform width between the rotor and the walls of the chamber.

A method of mixing one or more liquids is also provided. The method includes flowing the liquids to a mixing region between a static outer member and a rotatable inner member. The inner member has an outer surface and a cylindrical central portion. The mixing region has a cylindrical central portion, a substantially

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conical end portion, and a uniform width throughout. The liquids are mixed by rotating the inner member at a rotational speed sufficient to induce and maintain Taylor vortices in the mixing region while preventing the formation of bubbles in the mixing liquids in the mixing region. The mixed liquids are then discharged from the mixing region.

Brief Description of the Drawings

10 The invention can be described in greater detail with the help of the accompanying drawings.

FIG. 1 is a cross-sectional view of the apparatus of the present invention.

15 FIG. 2 is an alternative embodiment of the apparatus of the present invention which includes a fluid injection ring.

FIG. 3 is an alternative embodiment of the apparatus of the present invention which includes a cooling jacket.

20 FIG. 4 is an alternative embodiment of the present invention which includes a dye injection port.

FIG. 5 is a schematic depiction of the formation of Taylor vortices in an annular mixing region.

25 FIG. 6 is a cross-sectional view of an injection ring for use with the present invention.

FIG. 7 is an exploded view of a port in the injection ring of FIG. 6.

Detailed Description

30 In a preferred embodiment of the apparatus of the present invention, as depicted in FIG. 1, a rotor 2 is mounted concentrically with respect to a chamber 1 within a housing 6. Rotor 2 is attached to a rotatable shaft 8. Shaft 8 can be attached to a suitable rotating source,

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such as an AC motor (not shown), so that rotational speeds of 200-5000 RPM can be achieved. Rotor 2 extends longitudinally along axis 15. It has a central portion 11 and an end portion 13. Rotor 2 is cylindrical in shape at 5 its central portion 11. In a preferred embodiment, the surface of the rotor on both its central portion and its end is smooth and end portion 13 is tapered to allow air to be purged more effectively on start-up.

A first bulk solution inlet 10 provides conduit 10 means for introducing a liquid into chamber 1. Inlet 10 extends through the wall of housing 6 to an inlet-chamber 5. A second such conduit means is provided by bulk solution inlet 12, which also extends through the wall of housing 6 and thereby provides means for 15 delivering a second fluid to the chamber 1 via inlet-chamber 5.

After introduction into the inlet-chamber 5, the fluid components to be mixed are flowed to mixing region 4 of chamber 1 for mixing. The mixing region 4 is defined 20 by an annular gap between the central portions 7 and 11 of the chamber wall and the rotor, respectively, and has length L as shown in FIG. 1. Mixing is accomplished by rotation of rotor 2. After mixing, the mixed fluids are withdrawn through another conduit means, outlet 17.

25 Mixing can be optimized by choosing an appropriate rotational velocity of rotor 2, a suitable mixing region annular gap and length and a suitable rotor diameter for the combined viscosity and combined density of the materials to be mixed. Appropriate values are 30 selected so that the Taylor vortices (to be discussed below) form in the mixing region 4 during mixing, but without bubble flashing. To achieve these results, it has been found by experimentation that a Taylor number of between 125 and 340, inclusive, must be maintained in the 35 mixing region.

The Taylor number is a quantifiable measure of the mixing phenomena occurring in mixing chamber 4. The Taylor number can be expressed as follows:

5 $Ta = (2\pi R_i N d / 60 v) (d/R_i)^{1/2}$

where R_i is the radius of the cylindrical central portion of rotor 2; d is the distance from the outer wall of the cylindrical central portion of rotor 2 to the central wall 10 of chamber 1 in the mixing region 4; v is the kinematic viscosity of the resulting mixed fluids; and N is the inner cylinder (rotor) speed in RPM. Thus, for a given mixture's kinematic viscosity, peripheral velocity (governed by rotor speed), gap width and rotor radius must 15 be chosen so that a Taylor number from 125 to 340 is maintained. The values chosen for these parameters in the mixing of photographic emulsions are discussed below.

It has been found that extremely favorable mixing conditions can be obtained when flow instability 20 (formation of Taylor vortices) is achieved in the mixing region 4. Although flow is unstable, flow remains laminar, i.e., flow stream lines remain distinct from one another, and the liquids undergo laminar mixing as long as the Taylor number in the mixing region remains below 400. 25 Turbulent flow can be expected to occur above this Taylor number. When the point of instability is reached, the fluids being mixed begin to form a series of vortices whose axes are located along the circumference of the rotor and which rotate in alternately opposite directions. 30 as shown in FIG. 5. The formation of these vortices is desirable for mixing because it increases the surface area of the interface between the fluids being mixed, and hence liquids can be mixed efficiently for a given power requirement.

In addition to more efficient mixing, the formation of Taylor vortices also results in more uniform mixing of the fluids. Mixing uniformity is enhanced when a uniform velocity profile is achieved between the inner 5 and outer cylinders. This ensures that all fluid elements between the cylinders see the same shear forces and in turn achieve the same degree of mixing. These shear forces are optimized in the presence of Taylor vortices. The formation of Taylor vortices provides more uniform 10 shear action and, hence, more uniform mixing throughout the entire mixing region. A Taylor number of at least 125 is necessary to produce the uniformity required to mix photographic materials.

While maintaining the Taylor number above a 15 minimum value is desirable to promote uniform mixing, achieving a Taylor number that is too high will result in bubble flashing in the mixing region. Bubble flashing occurs when dissolved air is driven out of solution by high localized pressure differentials. Bubble flashing is 20 undesirable because the presence of bubbles in photographic emulsions can result in defects in the subsequently-coated photographic film. To prevent bubble flashing in photographic emulsions, it is necessary that the Taylor number be no greater than 340.

25 The parameter values selected to achieve the desired Taylor number were chosen so as to minimize component cost and material waste, while at the same time maximizing mixing efficiency and uniformity. Thus, the preferred rotor speed chosen was 1725 RPM, so that the 30 mixer can be used with many standard, inexpensive AC motors. This rotor speed allows selection of gap and rotor dimensions which result in a minimum amount of material waste during cleaning.

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In accordance with the present invention, the gap is relatively smaller than that which has heretofore been used. It has been found through experimentation that good mixing quality is achieved when the gap size is chosen so that the ratio of gap width to the central radius of rotor 2 (d/r) is about 0.1 to just below 0.2. This d/r ratio is critical for mixing liquids having a combined viscosity which is moderate, i.e., within the range from about 5×10^{-5} to about $6 \times 10^{-3} \text{ m}^2/\text{s}$. The mixing of liquids whose combined viscosity falls within this range is one of the contemplated uses of the present invention. Apparatus having a d/r ratio of 0.2 or more are unsuitable for use in mixing photographic emulsions. In a mixing structure with a small gap width relative to rotor radius, i.e., with a d/r ratio of 0.1 to just below 0.2 and using fluids with viscosities lying within the range from about 5×10^{-5} to about $6 \times 10^{-3} \text{ m}^2/\text{s}$ and a rotational velocity of 1725 R.P.M., the resulting vortex-like motion of the fluid components produces effective and high-quality mixing.

As discussed previously, in the mixing of liquid photographic materials, it is desired to maintain a constant constituent residence time in the mixer throughout the mixing process. To enhance uniform residence time, the apparatus and method of the present invention are calculated to maintain plug flow through the mixing region. That is to say, as a unit of fluid is introduced into the mixing region, the increased pressure in the mixer as a result of introducing this unit causes an identical volume of mixed fluid to be displaced from the mixer. Thus, fluid flows through the mixing region in a first-in first-out manner, i.e., a particular unit of fluid is not discharged from the mixer later than a unit of fluid introduced into the mixing region after that unit.

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Plug flow conditions are effected by providing a uniform gap width between the rotor 2 and the inner wall 7 of the housing's chamber 1 in the mixing region 4. In this way, a uniform cross-sectional area along the 5 longitudinal axis 15 of rotor 2 can be achieved throughout the mixing region 4. This uniform cross-sectional area is also maintained in chamber 1 downstream of mixing region 4, including outlet 17. Because the cross-sectional area is uniform, there are no dead zones 10 in which mixed liquids can collect and thereby remain in the mixing region for an unacceptably long time. Plug flow also has the advantage of allowing quick purging of air and other fluids from the mixing region during start-up and cleaning. Furthermore, plug flow insures a 15 suitable back mixing ratio in the mixer. As discussed previously, a low back mixing ratio is desirable, and the geometry of the mixing chamber in the present invention provides a ratio of just 1.08. The geometry of the mixing chamber is designed to minimize dead zones and maintain 20 aspect ratios of one where required, the aspect ratio being defined as the ratio of height to length of a given fluid flow volume.

In a preferred embodiment of the present invention, the rotor diameter is about 66.7 mm and the 25 mixing region gap width is about 4.8 mm. The selection of these parameters provides several advantages. The gap width is small enough to minimize hold-up volume to a value which minimizes material waste during mixer cleaning. This rotor diameter yields a d/r ratio within 30 the acceptable range for the mixing of photographic materials. Finally, selection of the gap and diameter values as stated above allows a standard, 1725 RPM AC motor to be used to drive rotor 2 and still maintain a Taylor number between 125-340 for the viscosities of the

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solutions mixed. Alternative embodiments of the present invention use a rotor diameter of either about 44.4 mm with a gap width of about 3.2 mm.

The mixer and method of the present invention were designed for use in photographic emulsion delivery systems. In such systems, pressure pulsations can cause non-uniform delivery of product, which can adversely affect the emulsion uniformity required on photographic films. To prevent such pressure pulsations, a preferred embodiment of the present invention uses a rotor having a smooth outer surface on both its central wall and conical end wall.

As shown in FIG. 1, means for injecting a fluid into the mixing region 4 can be provided in a preferred embodiment of the present invention. This means is provided by an injection port 14. The injection port 14 allows introduction of such constituents as catalyzing agents or reactants. The desired fluid velocity through this port is 1 m/sec, with a minimum velocity requirement of .33 m/sec. The advantage obtained from introducing such components directly into the mixing region in this manner is that they are rapidly dispersed.

In an alternative embodiment of the present invention, as shown in FIG. 2, means for injecting a fluid into the mixing region is provided by an injection ring 16. The injection ring 16, which is attached to the exterior wall of chamber 1, is used primarily for introducing a high solids slurry into the dispersing binder in the mixing region. Such a method of introduction is useful because complete mixing can be attained quickly. The injection ring includes anywhere from five to twenty ports 9 through which the slurry can be introduced and quickly dispersed throughout the fluids in the mixing region. Alternatively, substances such as

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dyes, catalyzing agents or water can be injected into the mixed liquids in this manner.

As seen in FIGS. 6 and 7, the ports 9 are tapered to provide optimum dispersal of solution into the mixing region. As can be seen, fluid enters port chamber 22, and then enters the mixing region through nozzle 24. Nozzle 24 causes the injected fluid to be dispersed and to enter the mixing region at a high velocity. The desired fluid velocity through these ports is 1 m/sec, with a minimum velocity requirement of .33 m/sec.

An alternative embodiment of the apparatus and method of the present invention can be used to mix relatively high viscosity liquids to produce a homogeneous, non-disperse product. Such an embodiment of the present invention is particularly useful for such purposes because it is capable of producing a homogeneous emulsion having dispersion droplets milled down to a desirable size. In this embodiment of the invention, shown in FIG. 3, only a single bulk solution inlet 10 is provided.

An alternative embodiment is shown in FIG. 4. It has a cooling jacket 26 to dissipate heat and is useful for mixing high viscosity solutions which require increased power. The preferred configuration in this embodiment uses a 4.8 mm gap with a 71 mm rotor or a 6.4 mm gap with a 95 mm rotor. The rotor speed required for these configurations ranges from about 200-1,000 RPM. Thus, this configuration, too, can be used with a small AC motor.

Alternatively, the apparatus of the present invention can be used for the injection of dyes into a high kinematic viscosity (0.05 to 0.35 m²/S) solution being mixed, as also shown in FIG. 4. The dyes are injected via dye injection port 18. This configuration

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uses a gap of either about 4.8 mm or 6.4 mm and a rotor diameter of about 71 mm. Rotor speed can vary between about 200-1,000 RPM.

The invention has been described in detail with particular reference to the preferred embodiments thereof, but it will be understood that variations and modifications can be effected within the spirit and scope of the invention.

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We Claim:

1. An apparatus for mixing one or more liquids, comprising:
 - 5 a stationary chamber having a substantially cylindrical central wall and an end wall; and a rotor positioned within said chamber and comprising a substantially cylindrical central portion, an end portion, a central radius, a longitudinal axis, and 10 having an outer surface extending along the cylindrical and end portions, said surface being spaced from the walls of the chamber and defining a mixing region of uniform width between the rotor and said substantially cylindrical central wall of said chamber.
- 15 2. The mixing apparatus according to claim 1, wherein said mixing region has a uniform cross-sectional area along said axis.
- 20 3. The mixing apparatus according to claim 2, wherein said outer surface of said rotor is smooth.
4. The mixing apparatus according to claim 1, wherein the ratio of said mixing region width to said 25 central radius is from about 0.05 to less than 0.2.
5. The mixing apparatus according to claim 4, wherein said ratio is about 0.1.
- 30 6. The mixing apparatus according to claim 1, wherein said uniform width is from about 3 to 5 mm.
7. The mixing apparatus according to claim 1, further comprising:

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first conduit means attached to said chamber for introducing said liquids into said chamber; and second conduit means attached to said chamber for withdrawing a mixture of said liquids from said chamber.

5

8. The mixing apparatus according to claim 1 further comprising means for rotating the rotor within said chamber.

10

9. The mixing apparatus according to claim 1 further comprising cooling means attached to said stationary chamber for cooling said mixing apparatus.

15

10. The mixing apparatus according to claim 1 wherein said end wall and said end portion taper.

11. The mixing apparatus according to claim 1 further comprising injection means for injecting liquids into said mixing region.

20

12. The mixing apparatus according to claim 11 wherein said injection means is an injection ring having at least one injection port, whereby said liquid to be injected can be injected at said at least one injection port within said mixing region.

13. The mixing apparatus according to claim 1 further comprising a geometry conducive to plug flow.

30

14. A method of mixing one or more liquids, comprising:

introducing said liquids to an annular mixing region between a static outer member and a rotatable inner member, said annular region having a cylindrical central

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portion, an end portion, and a uniform cross-sectional area;

mixing said liquids by rotating said inner member at a peripheral velocity; and

5 discharging said mixed liquids from said mixing region.

15. The method according to claim 14 wherein said rotatable inner member has a smooth outer surface.

10 16. The method according to claim 14 wherein said peripheral velocity is sufficient to induce and maintain Taylor vortices in said mixing region while preventing the formation of bubbles in the mixing liquids
15 in said mixing region.

17. The method according to claim 14 wherein the Taylor number, T_a , in said mixing region is maintained below about 340, said Taylor number being defined as

20
$$T_a = (2\pi R_i N d / 60 \nu) (d/R_i)^{1/2}$$

where N is the rotational speed of said inner member, d is said uniform width, R_i is the radius of said inner member
25 at its said central portion, and ν is the kinematic viscosity of said mixed liquids.

18. The method according to claim 17 wherein D is from about 3 to 5 mm.

30 19. The method according to claim 17 wherein N is maintained in the range of approximately 200 to 5000 R.P.M

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20. The method according to claim 14 wherein N
is maintained at approximately 1000 to 2500 R.P.M.

21. The method according to claim 17 wherein
5 said Taylor number in said mixing region is at least about
125.

22. The method according to claim 17 wherein
said Taylor number is maintained above about 125.

10

23. The method according to claim 14 further
comprising maintaining said liquids in said mixing region
for a constant residence time.

15

24. The method according to claim 14 further
comprising maintaining laminar flow of said liquids in
said mixing region.

25. The method according to claim 14 further
20 comprising injecting a liquid material into said mixing
region after the induction of said Taylor vortices.

26. The method according to claim 14 further
comprising maintaining plug flow in said mixing region.

25

27. An apparatus for mixing at least one liquid,
comprising:

a stationary chamber having a substantially
cylindrical central wall and a substantially conical end
30 wall;

a rotor positioned within said chamber and
comprising a substantially cylindrical central portion, a
substantially conical end portion, a central radius, a
longitudinal axis, and having a smooth outer surface

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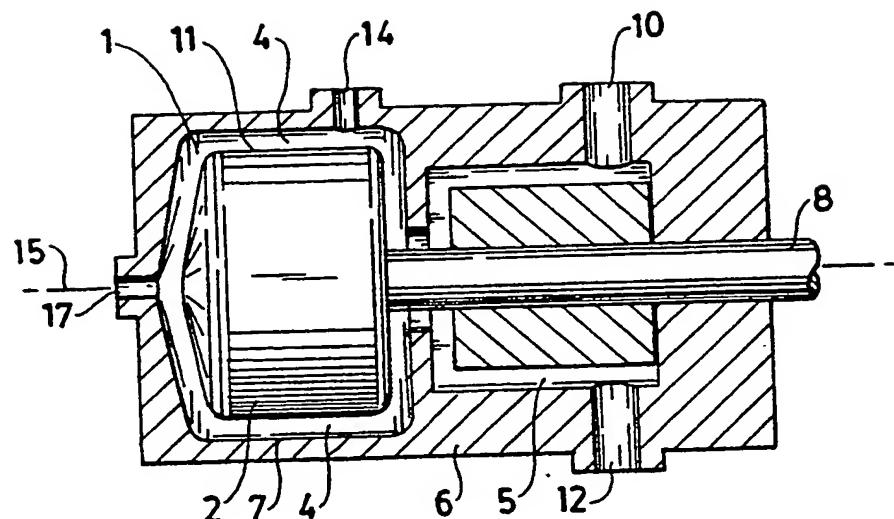
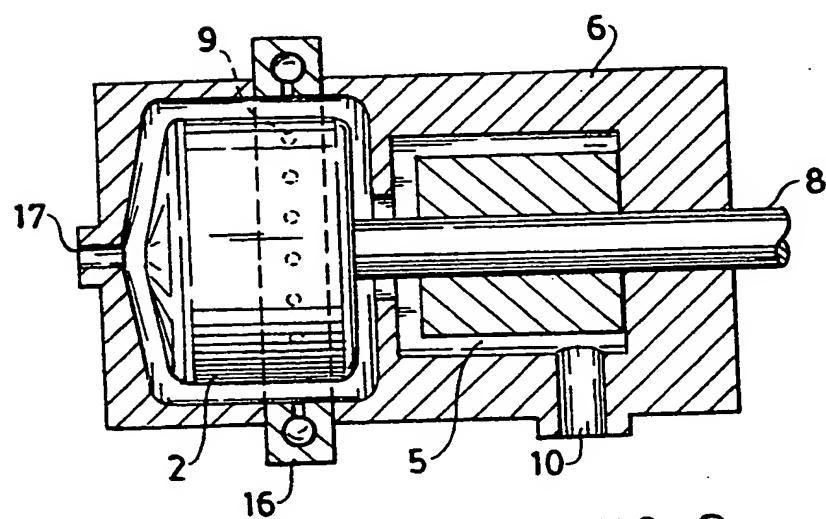
spaced from the walls of the chamber and defining a mixing region of uniform width between the rotor and the walls of said chamber;

inlet means for introducing said one or more liquids;

injection means for injecting a liquid material into said mixing region; and

a geometry conducive to plug flow.

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FIG. 1FIG. 2

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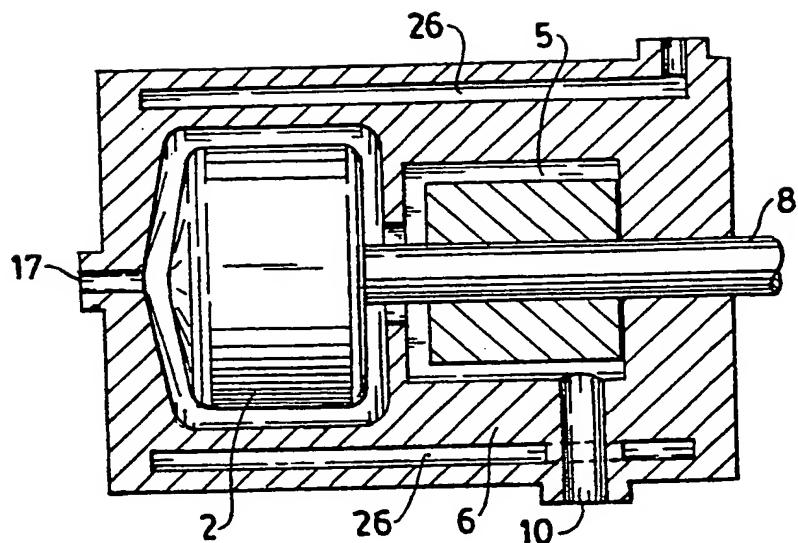


FIG. 3

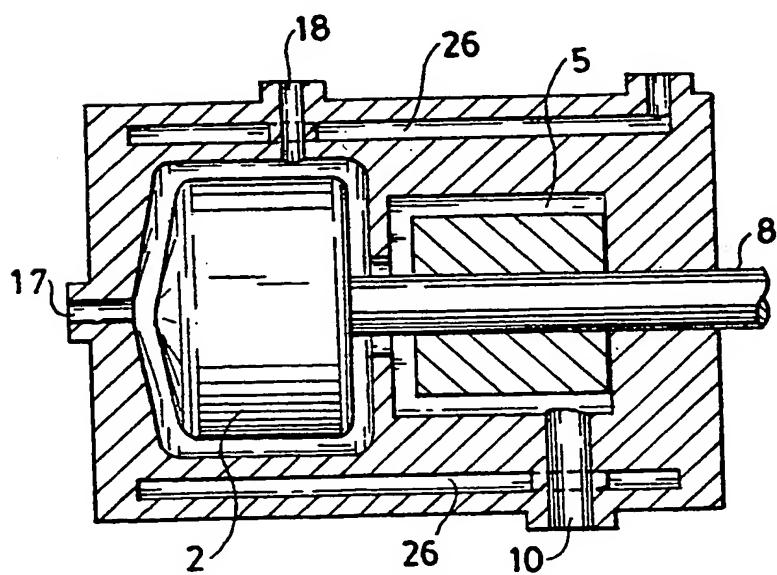
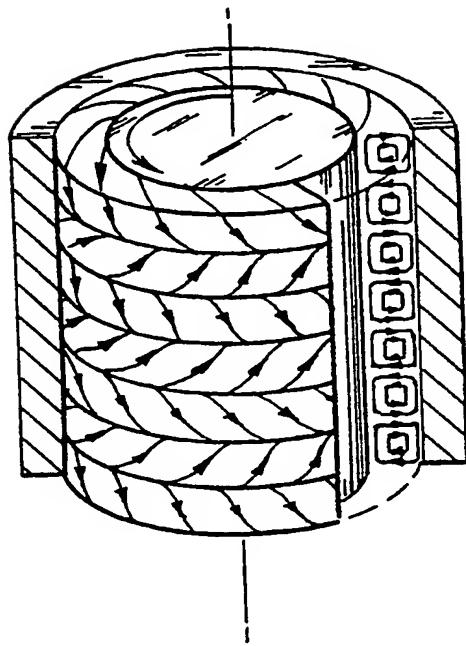
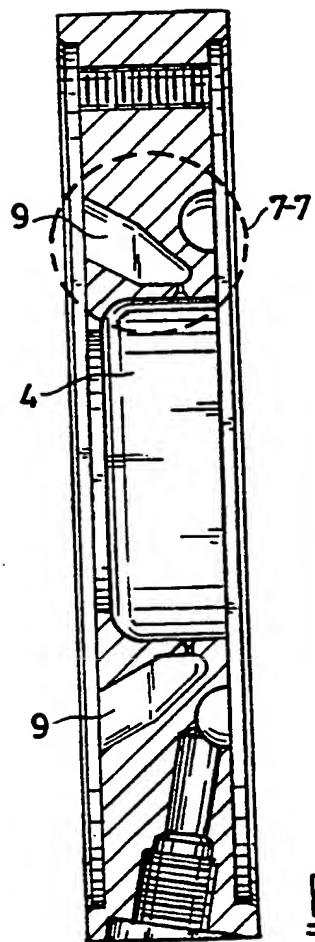
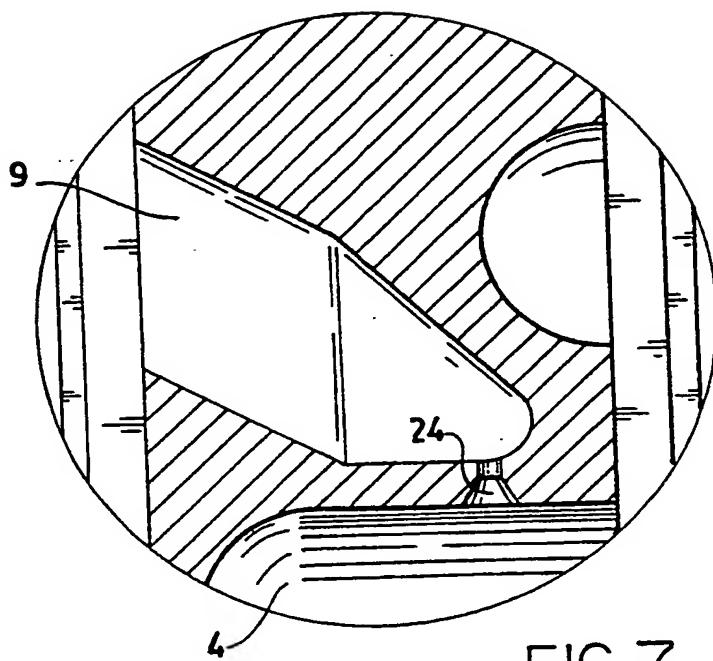


FIG. 4

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FIG.5FIG.6FIG.7

INTERNATIONAL SEARCH REPORT

PCT/US 92/09695

International Application No

I. CLASSIFICATION OF SUBJECT MATTER (if several classification symbols apply, indicate all)⁶

According to International Patent Classification (IPC) or to both National Classification and IPC
 Int.Cl. 5 B01F5/06

II. FIELDS SEARCHED

Minimum Documentation Searched⁷

Classification System	Classification Symbols
Int.Cl. 5	B01F

Documentation Searched other than Minimum Documentation
 to the Extent that such Documents are included in the Fields Searched⁸

III. DOCUMENTS CONSIDERED TO BE RELEVANT⁹

Category ¹⁰	Citation of Document, ¹¹ with indication, where appropriate, of the relevant passages ¹²	Relevant to Claim No. ¹³
X	US,A,4 174 907 (NAM P. SUH) 20 November 1979 see figure 3 ----	1-27
X	DATABASE WPI Week 7813, Derwent Publications Ltd., London, GB; AN 78-24700 & SU,A,508 262 (PARFENOV) 5 July 1977 see abstract ----	1-27
A	US,A,3 127 152 (SCHRENK) 31 March 1964 ----	9,11
A	EP,A,0 277 660 (MEMBREX) 10 August 1988 ----	
A	EP,A,0 486 974 (FUJI) 27 May 1992 ----	-/-

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IV. CERTIFICATION

Date of the Actual Completion of the International Search 19 JULY 1993	Date of Mailing of this International Search Report 04.08.93
International Searching Authority EUROPEAN PATENT OFFICE	Signature of Authorized Officer PEETERS S.

III. DOCUMENTS CONSIDERED TO BE RELEVANT (CONTINUED FROM THE SECOND SHEET)		
Category	Citation of Document, with indication, where appropriate, of the relevant passages	Relevant to Claims No.
A	US,A,1 721 121 (JENSEN) 16 July 1929 -----	
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